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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of Marc-Olivier Coppens

Serial No. 09/801,971

Examiner:

G.P. Straub

Filed: April 29, 1999

Art Unit:

1754

For:

METHOD FOR OPERATING A CHEMICAL AND/OR PHYSICAL PROCESS BY MEANS OF A HIERARCHICAL FLUID INJECTION SYSTEM

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DECLARATION

The Assistant Commissioner of Patents Washington, D.C. 20231

Dear Sir:

The undersigned, Marc-Olivier Coppens of Tuinwijklaan 47, B-9000 Gent, Belgium, herewith declares as follows:

- I am Marc-Olivier Coppens inventor of the above-referenced patent application.
- 2. I am currently associate professor in chemical engineering at Delft University of Technology (T.U. Delft), the Netherlands. For about 10 years I have been involved in research on chemical engineering. Before taking up a position at Delft University of Technology, I have been lecturing and researching at the University of California, Berkeley, at Yale University, and at the University of Gent, Belgium, where I obtained my Ph.D. My specialization within the field of chemical engineering is in modeling and control of the influence of geometry on physico-chemical phenomena of industrial processes, in particular of multi-phase processes, such as fluidized beds and bubble columns. I am author and co-author of numerous publications on these topics, as is evidenced by the enclosed publication list.

- 4. The ordinarily skilled person realizes that most multi-phase processes, such as fluidized beds or bubble columns, involve turbulence (i.e., turbulent agitation, see, for example, the enclosed copy taken from Kirk-Othmer's Encyclopedia of Chemical Technology, third edition, 10 (1984) p. 557). The mixing device, described by Kearney et al. in Fractals in Engineering, uses laminar flows to obtain mixing without turbulence. The mixing that is the object of Kearney et al., implies the use of easily miscible fluids, i.e., fluids that form a single phase. Therefore, the skilled person would have expected Kearney's mixing device to be useful only for single-phase laminar processes. To apply these devices as injectors in multi-phase processes, which processes normally and by their nature do not involve laminar flow, is therefore not obvious. Also, the improvement of the operation of multi-phase processes, which may be obtained with the injectors of my invention, is in no way derivable from the publication of Kearney, nor from any other.
- 5. The ordinarily skilled person would know that scaling up multi-phase processes (e.g., from laboratory scale to industrial scale) is difficult or impossible, particularly if it is attempted to maintain the same hydrodynamics. Such scale-up has depended up till now, at least in part, and often entirely, on empirical - viz. theoretically not (fully) understood - approaches, and usually involves cumbersome experiments in which the size of the equipment is increased in several steps, before an industrial scaled plant can be designed and constructed. As a result, significant errors have arisen in the past in such scaleup efforts, inter alia due to the effect of hydrodynamics in multi-phase processes. It is stressed that such errors may have major implications, since equipment on an industrial scale requires considerable capital investments, while the economic viability of the processes in which this multi-phase equipment is used depends strongly on the performance of this equipment, which often forms the heart of such processes. Unsuccessful scale-up may give rise to bad performance, e.g., conversions that are lower than expected, and may thus jeopardize the economic viability of the process. For this reason, considerable effort has been given in the past to develop models that predict the behavior of multi-phase processes. Up

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until now, however, these efforts have not lead to a full understanding of the processes, and scale-up of multi-phase processes remains a serious challenge. The present invention provides for a considerable contribution with respect to scaling up, since it provides for a different approach in which information obtained from scale experiments using the injectors of the invention is used to obtain large scale equipment, having the same hydrodynamics. Surprisingly, this may be done simply by keeping the parameters N, D and Δ the same for the small scale and large scale process. Since the cited publications are completely silent with respect to such an approach, the skilled person would not have expected, based on the cited documents, that a method of scaling up physical and chemical processes would be possible in a predictable manner, once N, D, and Δ are determined in a small-scale vessel, as is done in accordance with the present invention.

6. The undersigned declares that all statements made herein are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements and the like so made may jeopardize the validity of the document, or application, or any patent issuing thereon.

Signed this 24th day of January, 2001

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Dr. Marc-Olivier Coppens

List of publications of Marc-Olivier Coppens:

1. M.-O. Coppens and G. F. Froment, 1995, Diffusion and roaction in a fractal catalyst pore - I. Geometrical aspects. *Chem. Engng Sci.* 50, 1013-1026.

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- 4. M.-O. Coppens and G. F. Froment, 1995, Knudsen diffusion in porous catalysts with a fractal internal surface. Fractals 3, 807-820.

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FLUIDIZATION

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ADVANTAGES AND DISADVANTAGES OF THE FLUIDIZATION TECHNIQUE

Advantages

Temperature Control. The ability of the fluidized-solid bed to approach isothermal conditions is the outstanding advantage of this method over other methods of carrying out reactions. Of the several reaction variables, temperature is one of the most important, for reaction rates change exponentially with temperature (often doubling for a 10°C change). Commonly there are several competing reactions in which a temperature change of a few degrees may shift the balance of the several rates.

Three factors, in order of their importance, of the relatively close control of temperature in a fluidized-solid bed are:

(1) Turbulent agitation within the fluidized mass, which breaks and disperses any hot or cold apots throughout the bed before they grow to significant size. The catalytic activity differs somewhat for each particle, and those with greater activity accelerate the reaction in their neighborhood to a greater extent; consequently, their temperature is different than that of the surrounding particles of lower activity.

(2) High heat capacity of the bed is relative to the gas within it. This factor stabilizes the temperature of the bed, permitting it to absorb relatively large heat surges with only small temperature changes. For example, a bed of ordinary sand, fluidized with air at a solids concentration of ca 1120 kg/m⁹ (70 lb/ft³) would contain only about

 0.8 kg air/m^3 (0.05 lb air/ft³), corresponding to a mass ratio of ca 1400:1.

(3) A high-heat-transfer rate, which is possible because of the large amount of transfer surface per unit volume of the fluidized bed. This permits rapid leveling of any temperature surges either from the incoming gas or from reactions within the bed. Although the heat transfer coefficients are not unusually high, the amount of surface per unit volume is very large; eg. the surface area of a bed of ordinary sand would be in the range of $3280-16,400 \text{ m}^2/\text{m}^3$ ($1000-5000 \text{ ft}^2/\text{ft}^3$) of bed.

The point-to-point variation of temperature in large fluidized-catalyst beds is less than 10°C when the feed-gas temperature is not greatly different from that of the bed, and particularly if the inlet gas is carefully distributed.

Continuity of Operation. The ability to handle the fluidized solid like a liquid permits the technique to be easily adapted to many continuous operations, thereby obtaining the advantages of lower labor requirements, precise and automatic control of process variables, and uniformity of product quality.

Heat Transfer. The fluidized-solid technique is a convenient method for transferring heat, either alone or in conjunction with other operations, such as catalysis, gas-solid reactions, and transport of solids and fluids incidental to these operations

Catalysis. The fluidized-solid technique can be used for the contact of free flowing, nonsticky, granular solids with gases. It may be applied in catalytic gas reactions in which solid catalysts are used. The technique has been used most widely for the catalytic cracking of petroleum because of: (1) control of reaction temperature; (2) maintenance of uniform catalyst activity (as well as continuous catalyst regeneration); (3) continuous removal of solid by-products; (4) supply of heat to endothermic reactions; (5) simple equipment with few moving parts; and (6) continuous operation with automatic control.